Comment on: "Auger decay, Spin-exchange, and their connection to Bose-Einstein condensation of excitons in Cu_2O " by G.M. Kavoulakis, A. Mysyrowicz (cond-mat/0001438)

A. B. Kuklov

Dept. of Eng. and Physics, College of Staten Island, CUNY

In the very recent work [1] a new mechanism of the interconversion of the triplet excitons into singlet excitons in Cu_2O has been suggested. In accordance with it, two triplet excitons with the opposite (internal) angular momenta may collide and interconvert into a pair of the singlet excitons. Estimates presented in [1] show that such an interconversion is the most effective channel for the decrease of the triplet exciton density. This questions the commonly accepted view that the Auger decay is the primary channel for the decay (see in [2]). Furthermore, it has been pointed out in [1] that the actual rate of the Auger decay must be several orders of magnitude less than it was previously calculated.

In this comment, it is suggested that the mechanism [1] leads to a verifiable prediction about the rate of the decrease of the triplet exciton density as function of the polarization of the incident laser light inducing the two-photon creation of the triplet excitons. As a matter of fact, this rate can be greatly reduced if the polarization of the laser light is properly chosen.

First, it is worth noting that a necessary condition for the triplet-singlet interconversion [1] is that before the collision a pair of the triplet excitons has total angular momentum $J = 0$, that is, they form a singlet state (invariant under the point group rotations). Thus, the rate of the interconversion should be extremely sensitive to the initial state of the excitonic ensemble. If this state is a thermal mixture with random orientation of the excitonic spins, then each exciton can easily find another one with the opposite J_z, so that the collision between them would result in a pair of the singlet excitons [1]. On the contrary, if initially excitonic spins were aligned, the collision induced interconversion will be completely suppressed because of the conservation of the angular momentum of the colliding pairs. Thus, aligning angular momenta of the triplet excitons in one way or another should prevent the triplet excitons from transforming into the singlet excitons in accordance with the mechanism [1]. This property can be used as a test for the mechanism [1].

One way for preparing triplet excitons with preferential orientation of their spins is a direct creation of the coherent triplet excitons employed in [3]. In this work, the triplet excitons have been created by the two-photon direct transitions: The incoming laser field was tuned to the half of the excitonic frequency, so that the two-photon transition was in exact resonance with the triplet excitons. Such a method allows to create a dense and coherent cloud of the triplet excitons. This is practically a direct mean of creating a condensate of the triplet excitons. However, fast collision induced decay of the triplet excitons may destroy this coherence on the scale of few nanoseconds. On one hand, this is the case if the Auger decay is responsible for the triplet exciton depletion because this channel is not sensitive to the orientation of the angular momenta of the colliding pairs. On the other hand, if the primary channel for the decay is the mechanism [1], it should be possible to use such a polarization of the cloud that the created condensate of the triplet excitons is stable on much longer time scale.

It is possible to employ general symmetry considerations, and make a suggestion for the choice of the orientation of the incoming laser fields in the geometry of the experiment [3]. Indeed, the two-photon process of creation of the triplet exciton corresponds to the interaction term in the energy density

$$H_{le} = \sum_{a=\pm 1,0;i,j} \psi^\dagger_{(a)} Q^{(a)}_{ij} E_i E_j + H.c. \qquad (1)$$

where $\psi_{(a)}$ stands for the triplet exciton Bose field which has three projections $a = \pm 1$ and $a = 0$ of the (internal) angular momenta; E_j denotes three space components of the incoming laser field $\sim E_j \exp(-i\omega t)$ which is taken in the rotating wave approximation; $Q^{(a)}_{ij}$ are corresponding matrices representing the point symmetry (including spins) of Cu_2O in such a way that (1) is invariant under this symmetry. The interaction term responsible for the decay [1] can be represented in the contact form (S-wave channel) as

$$H_{op} = g_{op}\psi^\dagger\psi^\dagger(\psi_{(+1)}\psi_{(-1)} + \psi_{(-1)}\psi_{(+1)} + \\ +\psi_{(0)}\psi_{(0)}) + H.c. \qquad (2)$$

where ψ is the field of the singlet excitons, and g_{op} is the interaction constant such that the rate estimated in [1] is $\sim g^2_{op}$. It is worth noting that the term in the brackets in (2) is invariant under the symmetry group (including spins) of Cu_2O, where the excitonic states are formed on the total angular momenta states of Cu (see in [4]). If the triplet excitons are created in such a manner that this invariant is zero, the interconversion process will be suppressed.

The induced fields $\psi_{(a)}$ are given from (1) as $\psi_{(a)} \sim \sum_{i,j} Q^{(a)}_{ij} E_i E_j$. If substituted into (2), this will result in the term describing four-photon production of the singlet excitons which in general should be significant as long as the mechanism [1] is dominant, provided the density

$|\psi_{(a)}|^2 \sim |\sum_{i,j} Q_{ij}^{(a)} E_i E_j|^2$ of the induced triplet excitons is large enough. In fact, the symmetry of $Q_{ij}^{(a)}$ is the same as that of the tensors of the direct quadrupole transitions for the triplet excitons. Using this, it is possible to find the energy density (2) as

$$H_{op} \sim g_{op} \psi^\dagger \psi^\dagger (E_x^2 E_y^2 + E_x^2 E_z^2 + E_y^2 E_z^2) + H.c. \quad (3)$$

where E_x, E_y, E_z refer to the components of the laser field with respect to the principal cubic axes of Cu_2O. Accordingly, the requirement

$$E_x^{-2} + E_y^{-2} + E_z^{-2} = 0 \quad (4)$$

insures that the interconversion process [1] described by (2), (3) is zero in the dominant s-wave channel as long as no thermalization of the created triplet excitons occurs.

A solution of (4) for the six components of $E_j = E_j' + iE_j''$, where E_j' and E_j'' stand for the real and imaginary parts of E_j, respectively, can be represented as follows

$$E_j = \frac{1}{\epsilon_j' + i\epsilon_j''} \quad (5)$$

in terms of the two auxiliary real vectors ϵ_j', ϵ_j'' which are arbitrary except for the conditions

$$\sum_j \epsilon_j'^2 = \sum_j \epsilon_j''^2, \quad \sum_j \epsilon_j' \epsilon_j'' = 0. \quad (6)$$

The interpretation of these conditions is straightforward: the complex vector E_j represented by (5) should be chosen in such a way that the two auxiliary vectors ϵ_j' and ϵ_j'' are equal in magnitude to each other and are mutually orthogonal. For the case of the incidence of the light along the direction (1,1,1), the solution of (6), (5) gives that $\sum_j E_j' E_j'' = 0$ and $\sum_j E_j'^2 = \sum_j E''^2$. Note that, given this, the interconversion rate $k(T=0) = 0$, as opposed to the rate of the Auger decay which is not sensitive to temperature and the mutual orientation of the excitonic angular momenta of the colliding pairs.

At finite temperatures $T \neq 0$, the normal component - thermal triplet excitons - is present in addition to the condensate. This component should be characterized by zero net spin polarization due to the interaction with phonons. Thus, the interconversion process [1] will take place. Its rate $k(T)$ is proportional to the normal density n'. Thus, it must be strongly temperature dependent. It is straightforward to find an estimate for $k(T)$ in the temperature range $\mu(0) < T \ll T_0$, where $\mu(0) = 4\pi a n_0$ and a, n_0 stand for the excitonic scattering length and the exciton condensate (spin-polarized) density, respectively; and T_0 denotes the temperature of the excitonic Bose-Einstein condensation. Indeed, in this range the normal component behaves almost as an ideal gas. Thus, the estimate follows from Eqs.(8-10) of Ref. [1] where the total density is replaced by the density of the normal component $n' = n_0 (T/T_0)^{3/2}$ [5]. Accordingly, the ratio of the rate of the interconversion $k(T)$ at $T \neq 0$ for the spin-polarized excitonic condensate to the rate $1/\tau_{o,p}$ [1] estimated for the case of the non-polarized cloud is

$$k(T)\tau_{o,p} \approx \frac{n'}{n_0} = \left(\frac{T}{T_0}\right)^{3/2} \ll 1 \quad (7)$$

for the temperatures under consideration. At temperatures $T < \mu(0)$, further significant reduction of the rate should occur due to the interaction between the triplet exciton condensate and the normal component.

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